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REGIONAL-SCALE POLLUTANT TRANSPORT STUDIES IN THE NORTHEASTERN UNITED STATES

by

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16. ABSTRACT

Field studies were conducted in the northeastern United States during the summers of 1979 and 1980 to obtain data for development and validation of urban and regionalscale photochemical air quality simulation model and to study the physical and chemical processes important in long-range pollutant transport.

This program consisted of four major components: 1) Regional mapping of pollutants over the northeastern U.S. using in situ aircraft sampling, airborne lidar, and surface sampling sites. 2) Eulerian and Lagrangian aircraft sampling of urban plumes and interacting urban plumes from their source areas to distances where they blend with the regional background burden. 3) Removal of pollutants by dry deposition to the surface and venting by clouds from the mixed layer to a more stable cloud layer. 4) Deployment of Lagrangian atmospheric markers (tetroons) to track specific atmospheric air volumes for subsequent sampling and to obtain data for uncertainty analysis of diagnostic transport models. A brief overview of the program is presented with preliminary results of analyses for each of the above components.

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INTRODUCTION

There has been an effort in recent years to assess the importance to local regulatory strategies of regional-scale pollutant transport, particularly for secondary pollutants such as oxidants, nitrates, and sulfates. The U.S. Environmental Protection Agency (EPA) has established modeling and field programs to address regional-scale transport of both ambient oxidant and acidic materials.

The EPA program to address regional-scale transport of oxidants is summarized here. The focal point of the program is the development and application of the Regional Oxidant Model (ROM) as part of the Northeast Corridor Regional Modeling Project (NECRMP). The ROM (Lamb, 1983) is a regional-scale Eulerian transport and photochemistry simulation model that will be used to

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provide inflow boundary conditions of ozone and precursors for application of urban scale air quality simulation models to cities in the northeastern U.S. The ROM will also provide a regional perspective for assessing the impact of oxidant control strategies of individual cities. The model is formulated to treat those chemical and physical processes that are presently thought to affect photochemical oxidant concentrations over diurnal time scales (e.g., nocturnal wind shear, nighttime chemistry, and effects of clouds on perturbing the photochemical reactions and venting of pollutants from the boundary layer). The area being modeled is truly regional in scope, extending from western Ohio to the East Coast and from northern Virginia across southern Maine and Ontario (see Fig. 1).

Several field studies were conducted in support of NECRMP. These were the Northeast Regional Oxidant Study (NEROS) field programs and the Urban Field Studies. The NEROS field programs, conducted during August 1979 and from mid-July to mid-August 1980, consisted of three basic sampling scenarios: 1) Regional Air Mass Characterization (RAMC) sampling designed to obtain model initialization and evaluation data over the extent of the ROM grid; 2) Regional Atmospheric Lagrangian (REAL) experiments involving Lagrangian characterization of the photochemistry within an air mass segment marked by a tetroon; and 3) Regional Urban Plume Characterization (RUPC) studies designed to obtain data to characterize the evolution of the urban plume from its source area

to the point where it blends with the regional background ozone burden. In addition, Urban Field Studies were conducted in four major Northeast Corridor cities during the summer of 1980 to obtain data for application of urban scale models.

This paper provides a brief overview of the various field studies and resulting data bases. The primary sampling scenarios are described and some results that were analyzed for the purpose of data base preparation and evaluation for modeling exercises are summarized.

DESCRIPTION OF FIELD PROGRAM

Aircraft were the basic sampling platforms for all the field programs. Most aircraft were instrumented for in situ continuous sampling of nitric oxide (NO), total oxides of nitrogen (NOx), ozone, nephelometer scattering coefficient, ambient temperature, relative humidity or dew point, and for grab sampling for later analysis of hydrocarbon species. Five aircraft, based at three locations in the Northeast, were used in the 1979 program, which emphasized RAMC and REAL experiments coordinated from Raleigh, North Carolina. Thirteen aircraft were used in various components of the 1980 program. REAL experiments were coordinated from Columbus, Ohio; REAL and RUPC experiments were conducted from Columbus, Ohio and from Baltimore, Maryland. Urban Field Studies were conducted in Washington, DC; Baltimore, Maryland; New York, New York; and Boston, Massachusetts. A number of special

experiments, including studies on ozone deposition and cloud venting of ozone from the boundary layer, were conducted near Lancaster, Pennsylvania, in 1979 and Columbus, Ohio, in 1980. The NECRMP field program and scenarios are diagramed in Fig. 2.

NEROS RAMC Scenario

The Regional Air Mass Characterization (RAMC) scenario was designed to obtain data over the three-dimensional domain of the NEROS grid for evaluating the overall performance of the ROM. Aircraft were deployed to obtain air quality data over 24 to 48-h periods. Sampling was conducted throughout the diurnal cycle and included flights within the residual mixed layer at night and in the cloud layer above the mixed layer. Seven RAMC scenarios were conducted in 1979 and two in 1980 with varying degrees of success. Most RAMC aircraft sampling was conducted in a forecast Lagrangian mode, i.e., an initial volume of air was sampled near midday along the upwind edge of the ROM grid at different altitudes within and above the mixed layer. Subsequent sampling traverses were made at approximately 6-h intervals at the predicted downwind Lagrangian location of the air mass sampled on the original traverse. Thus each aircraft traverse provided evaluation data based on the same initial boundary conditions.

The frequency of National Weather Service upper-air soundings at stations in the vicinity of the NEROS grid was increased to 6-h intervals and one additional site was added during both 1979 and

1980 programs (see Fig.3). The expanded network provided a meteorological data base for the execution of the regional model and for sensitivity assessment of various diagnostic trajectory models to density of observations (see Clarke et al., 1983).

NEROS REAL/RUPC Scenarios

Regional Atmospheric Lagrangain (REAL) experiments were also conducted during 1979 and 1980. The scenario for these experiments involved the release and tracking of tetroons and subsequent Lagrangian pollutant sampling at the location of the tetroon. The tetroons were used to mark specific segments of polluted air masses, power plant plumes, and urban plumes. Each tetroon carried an instrument package weighing appproimately 2 kg and consisting of an aircraft transponder and pressure encoder. The location and altitude of the tetroon was identified by Federal Aviation Administration regional flight service centers. The spatial and temporal coordinate information were relayed to the NEROS field headquarters from which the sampling aircraft were deployed. Twelve of the tetroons were released and tracked; two in 1979 and ten in 1980. The tetroon trajectories are shown in Fig. 3. A case study of a REAL experiment conducted in conjunction with a Regional Urban Plume Characterization (RUPC) experiment is given by Clark and Clarke (1983).

RUPC experiments were conducted only during the 1980 field study. The RUPC scenario consisted of sampling within an urban

plume from its source area to the point where it blended into the regional background. The RUPC's were conducted from both Columbus, Ohio, and Baltimore, Maryland, and, as noted above, sometimes in conjunction with a REAL scenario, i.e., a tetroon was sometimes used to mark a segment of an urban plume.

Urban Field Studies

Intensive field measurements were obtained during the summer of 1980 in Washington, Baltimore, New York City, and Boston to acquire an ambient air quality and meteorological data base for application of the Airshed urban scale model (Reynolds and Reid, 1978) to these cities.

The Airshed model requires that boundary and initial conditions of pollutant concentration, wind, and temperature be specified in three dimensions for each day modeled. Verification of the model requires specification of pollutant concentrations for each predictive time step. These requirements led to the development of a monitoring program consisting of continuous pollutant and meteorological measurements at the surface, pollutant measurements aloft (via aircraft), and upper-air meteorological measurements. An overview of these studies is provided by Possiel and Freas (1982).

NECRMP DATA BASE

A comprehensive NECRMP data base will eventually be compiled. Presently, however, it is convenient to discuss the individual elements as separate data bases: NEROS 1979, NEROS 1980, and Urban Field Studies.

The NEROS 1979 data base consists of RAMC measurements from three aircraft and enhanced radiosonde observations. In addition, there were site-specific data from Lancaster, Pennsylvania, consisting of aircraft profiles of pollutant concentrations (primarily ozone), surface ozone deposition measurements, and aircraft measurements of surface and cloud fluxes of ozone. These data are archived by platform; i.e., there is an individual data tape for each platform.

The 1980 NEROS data base is organized separately for two cities: Columbus, Ohio, the headquarters for the field program, and Baltimore, Maryland. The Columbus data base consists of pollutant and meteorological measurements from eight aircraft and many surface sites for RAMC, REAL, and RUPC experiments. A special feature of this data base is indirect measurements of ozone from a NASA aircraft equipped with a dual-wavelength lidar. Aircraft ozone flux measurements, both at the surface and in the vicinity of clouds, are also a part of this data base along with data from several ancillary platforms and the enhanced radiosonde network (Fig. 3). An overview of the Columbus program with a

listing of platforms is given by Vaughan et al. (1982). This data base is not complete but is to be archived both by platform and chronologically, i.e., the data will also be organized by date.

The 1980 NEROS Baltimore data base consists of in situ pollutant and meteorological measurements from three aircraft and a mobile van. There are also pollutant data from an enhanced surface network and a tethered balloon system, and meteorological data from pilot balloons, radiosondes, and acoustic sounders (Possiel and Freas, 1982). This data base is archived by platform and chronologically for 22 experimental days.

The Urban Field Studies data base consists of pollutant and meteorological measurements via aircraft for 13 days near Washington, 19 days near New York City, and 15 days near Boston. There are also pollutant and meteorological data from an enhanced surface network within 100 km of each of the cities and upper-air meteorological measurements from special radiosondes, pibals, and acoustic sounder in and near the cities.

A major effort to sample hydrocarbon species was incorporated into the overall field program. Over 550 grab samples (one to three minutes in duration) were collected from aircraft, and 790 one-hour integrated samples were collected at fixed urban surface sites in Columbus, Baltimore, Washington, New York City, and Boston. The samples were analyzed by gas chromatography for concentrations of 113 species.

The hydrocarbon measurements for both years and all cities have been collated into a comprehensive data base, which includes supplemental information on the ambient concentrations of other pollutants measured by the in situ aircraft and at the surface monitoring sites.

NECRMP DATA ANALYSES

Application of the data for modeling requires some preliminary preparation and evaluation regarding the quality and availability of data from the various platforms. Mission summaries were prepared describing the experimental scenarios, meteorological synopses, and platform deployment of the various field programs. More detailed presentations and evaluations were carried out for potential candidate days (based on quantity and quality of data) for model evaluation runs. This often led to interesting analyses and conclusions, some of which are summarized here.

Regional-Scale Transport

The following discussion summarize a case study of the RAMC scenario for 3-4 August 1979. The analysis, which is based primarily on aircraft measurements, pertains to an air mass followed and sampled in a Lagrangian mode through a diurnal cycle as it was transported from Ohio to the East Coast. Details are given by Clarke and Ching (1983).

On 2 August a weak cold front passed through the Ohio Valley and moved to the East Coast where it became stationary for about 48 h. Boundary layer transport winds behind the cold front were generally westerly and initial conditions for Lagrangian air mass sampling (and upwind boundary conditions for the ROM) were established by aerial sampling along track A-B in Fig. 5 from 1151 to 1359 EST on 3 August. Diagnostic trajectories initialized at 1300 EST on 3 August at three points along track A-B are also shown in Fig. 5. Based on meteorological data and forecast trajectories obtained over the next 24 h, four subsequent aircraft traverses were made along tracks C-D, E-F, G-H, and I-J at the times indicated. The tracks were specified to approximate Lagrangian sampling of the air mass as it was transported to the East Coast.

The air quality and meteorological data for each aircraft transect were subjectively analyzed to describe the large-scale features along the transect. The analyzed ozone cross-section for the second transect (track C-D) is shown in Fig. 6. The track was the predicted 6-h displacement of the air mass sampled earlier along track A-B. The analysis depicts a plume with an ozone concentration near 100 ppb in the northern part of the transect and ozone concentrations near 80 ppb above the mixed layer in both the northern and southern parts of the track.

Isopleths of ozone concentration near the center of the mixed layer (or its nocturnal residual), extracted from cross sections

of all the transects, are shown in Fig. 5. The analysis represents a quasi-Lagrangian field in that changes in concentrations along a trajectory roughly represent the Lagrangian derivative rather than a spatial change. Ozone concentrations were generally high over the grid and and exceeded the primary 1-h average National Ambient Air Quality Standard for ozone (0.12 ppm) over a large area of western New York. A backward trajectory analysis suggested that the air mass over western New York at 1900 EST 04 August was over Chicago, Illinois, 36 h earlier and was transported over urban source areas in northern Indiana and northern Ohio.

The temporal gradients of ozone concentrations were consistent with those expected from production and depletion processes. Between 1300 and 1800 EST on 3 August (tracks A-B and C-D respectively in Fig. 5), ozone concentrations increased as would be expected from chemical production processes. The increase was largest in the northern portion of the grid, the region of highest precursor emissions (Clark, 1980). Concentrations generally decreased between 1800 and 0800 EST the following morning and then increased on the afternoon of 4 August (track I-J). Concentrations were generally lowest during the early morning transect (track G-H). During the late evening through early morning hours, surface concentrations of ozone were between 0 to 30 ppb over the extent of the grid. These data support the concept of ozone being transported aloft at night

while being depleted in the surface layer.

Transport of Ozone in Urban Plumes

On 14 August 1980, a segment of the Baltimore-Washington composite plume, marked by a tetroon, was tracked and sampled by aircraft for 400 km along the Northeast Corridor (Clark and Clarke, 1983). The tetroon trajectory and aircraft sampling tracks are shown in Fig. 7. The tetroon was released immediately downwind from Baltimore at 0945 EST and was transported about 11 h along the Northeast Corridor into southern Connecticut. Aircraft sampling was conducted upwind from Baltimore prior to the release of the tetroon and in the vicinity of the tetroon (Lagrangian sampling) at locations immediately downwind from Baltimore, 70 km downwind from Baltimore, immediately downwind from Philadelphia, and in southwestern Connecticut.

Aircraft sampling upwind of Baltimore established the existence of an air mass ozone concentration of about 70 ppb. This concentration was fairly extensive both within the Washington plume, in the mixed layer (about 600 m at that time), and above in the residual mixed layer from the previous afternoon which extended to about 1800 m. Background ozone concentrations measured above the residual mixed layer were about 40 ppb.

Fig. 8 illustrates the ozone and NOx traces at three flight levels immediately downwind from Baltimore between 0955 and 1041

EST. The mixing height was about 700 m. Below the mixing height, a broad NOx plume from Baltimore sources apparently scavenged much of the developing Washington ozone plume. Ozone concentrations outside the NOx plumes increased from 70 ppb upwind to between 80 and 90 ppb downwind from Baltimore. Ozone continued to develop in the composite Washington-Baltimore plume and reached 170 ppb northwest of Philadelphia by late afternoon.

Ozone concentrations on the last set of transects in southwestern Connecticut were significantly reduced from those measured northwest of Philadelphia. Near the surface, ozone was severely scavenged by NO emissions from New York which produced NOx concentrations approaching 90 ppb. A sampling traverse at 1100 m showed nearly uniform ozone concentrations of about 100 ppb--a resevoir transported throughout the night and which could potentially be mixed to the surface the following day.

Simulation of Regional Transport

The tetroon data from the REAL experiments combined with the radiosonde data provided a unique opportunity to evaluate trajectory models and to assess the effects of increasing the temporal density of the radiosonde observations on model performance. The tetroon trajectories shown in Fig. 4 and those for two other tetroon data sets were simulated by three different diagnostic trajectory models which differed significantly in sophistication and input data requirements (Clarke et al., 1983).

The trajectory models evaluated were 1) the National Oceanic and Atmospheric Administration, Air Resources Laboratory Atmospheric Transport and Dispersion (ATAD) model (Heffter, 1980), 2) the National Center for Atmospheric Research (NCAR) Isentropic model (Haggenson and Shapiro, 1979), and 3) the Center for Air Pollution Impact and Trend Analysis (CAPITA) Monte Carlo model (Patterson et al., 1981). The ATAD model was evaluated for both 6- and 12-h radiosonde data. The NCAR model used only the standard 12-h radiosonde data, and the CAPITA model used only midday surface wind speed and direction measurements.

Comparison of the results generated by these techniques did not reveal a clearly superior model. There was a tendency in all the models for the tetroon to be to the left of its diagnostic position by about 10% of the trajectory length with a standard deviation of about 25% of the trajectory length. Apparently the midday surface winds used in the CAPITA model describe the large-scale flow features about as well as the ATAD model using four radiosondes per day, suggesting that a significant portion of the random features of the flow must be associated with space and time scales smaller than the 6-h radiosonde network.

The standard deviation of the differences as a function of travel time between the REAL tetroons (Fig. 4) and diagnostic positions by the ATAD model using 6-h data are shown as σ_E in Fig. 9. Also shown are the standard deviations of the separation of tetroon triads σ_T (Allen et al., 1967) and plume standard

deviations σ_{ph} based on data presented by Carras and Williams (1981).

The curves representing the tetroon triad data and plume data are not greatly inconsistent and are suggested to represent plume spreading due to turbulence, wind shear, and mesoscale atmospheric processes. Diagnostic trajectories are idealized simulations of the mean flow. The variances of the difference between the diagnostic and tetroon trajectories are due primarily to the inability of the wind fields and models to adequately simulate the motion of the air parcel associated with the tetroon. Thus, the differences between the σ_ε curve and the σ_p and σ_T curves are an indication of the uncertainty associated with the simulation of the mean flow. Had the transport component of the wind been precisely simulated by the diagnostic trajectories, σ_ε should have been similar in magnitude to σ_p and σ_T. The differences increase with increasing travel time and σ_e is about five times larger than σ_p after about 12 h, a result similar to that obtained by Allen et al. (1967).

Vertical Transport of Ozone by Clouds

Clouds play an important role in transport and photochemistry of ozone and precursors. For example, they reflect incoming radiation important in photochemistry of the lower layers; they can transport pollutants out of the mixed layer; they provide a medium for liquid phase reactions; precipitating clouds can return

upper level material to the surface in downdrafts; and they can alter the strength of the synoptic scale vertical velocity field which in turn affects mixed layer growth rates.

An important component of the NEROS field program was to investigate the effect of vertical transport by clouds, hereafter referred to as venting, on the variability of ozone concentrations in the mixed layer. In order to study the net transport of ozone via the cumulus route, special aircraft flights were made in both NEROS 1979 and NEROS 1980. The aircraft was instrumented with a gust probe system complemented with an on board inertial navigation system, to yield data on both the mean and turbulent component of temperature, moisture, 3-D wind, and ozone. Flights were made both within and above the mixed layer and included penetration of convective cloud elements. Preliminary results and further details regarding the deployment of this aircraft have been reported by Ching et al. (1983).

Fig. 10 shows plots of the departures from the mean of a few of the variety of parameters measured during a cloud layer traverse in the 1979 NEROS study. The actual cloud penetrations are indicated at the top of the figure and are associated with positive anomalies in all of the parameters. The ozone anomaly is about 5 ppb and ozone concentrations within the clouds were similar to those within the mixed layer.

Urban areas and their plumes may exhibit large ozone

concentrations and therefore large differences between mixed and cloud layer values. Consequently, venting is expected to be a significant feature of the ozone budget of urban plumes. For example, a cloud layer flight over Philadelphia, Pennsylvania on 27 August 1979 showed ozone during cloud penetrations of greater than 100 ppb, 50 ppb above ambient levels. Ozone concentrations in the mixed layer below the flight traverse were about 90 to 150 ppb.

NEROS field programs have confirmed cloud venting as an important process in the mixed layer ozone budget. Pollutants vented above the mixed layer may be transported without significant dilution or chemical transformations and eventually entrained back into the mixed layer to impact surface concentrations.

Characterization of Ozone Episodes

As part of the process of selecting days for application of the RAM and urban-scale models, each high ozone day in 1980 is being evaluated in terms of 1) the magnitude and spatial gradient of ozone transported across the modeling domain and within urban plumes and 2) the occurrence and complexity of mesoscale meteorological phenomena within the Northeast Corridor along with the impact of these features on observed ozone concentrations. Following is a synopsis of this type of episode characterization analysis for 5 August 1980.

A high pressure system was centered over the southeastern U.S. and a warm front extended east-west across Massachusetts and northern New York. In general, clear skies persisted from morning throughout mid afternoon and maximum surface temperatures were between 32 and 34oC.

Obvious ozone plumes extended downwind of the major Corridor cities as evidenced from the isopleths of maximum 1-h ozone concentrations (see Fig. 11). Areas of high ozone concentrations were observed to the northeast of Washington, Baltimore, Philadelphia, and New York City urban areas consistent with the boundary layer wind flow. Near Boston, the peak surface ozone concentration, 159 ppb, was measured to the west of the city in an area of convergent low-level flow associated with the slow northward advancement of a warm front across southern New England. Ozone concentrations and the configuration of the urban plumes aloft (measured by aircraft) were similar, except near Boston. A peak ozone concentration of about 200 ppb was measured aloft 50 km to the northeast of Boston in a layer of southeasterly flow.

Ozone concentrations outside the area of direct impact of Corridor emmisions were fairly homogeneous at about 60 ppb. Within the Corridor, ozone concentrations were generally high and exhibited sharp gradients. The National Air Quality Standard (0.12 ppm) was exceeded over a large area and even upwind of most major urban areas. Thus it appears that a relatively high background ozone burden is supplementing strong ozone production

downwind of the major urban areas and these plumes subsequently impact other urban areas.

CONCLUDING REMARKS

Field programs were conducted during the summers of 1979 and 1980 in support of the development and evaluation of the ROM and for application of the Airshed model to cities in the Northeast Corridor. The field programs have resulted in an extensive data base of ambient pollutant concentrations and meteorological measurements for the northeastern United States. Some preliminary analyses for the purpose of preparation and evaluation of the data base for ROM modeling exercises have led to some interesting insights on horizontal and vertical transport processes of importance to regional-scale transport, e.g., the confirmation of convective cloud venting as an important process in the mixed layer ozone budget.

The data base is available for further analyses and model evaluation programs. It is most appropriate for research programs leading to enhanced understanding of physical and chemical processes by which ozone is produced and transported to regions both near and far from its source. Additional information on the data base can be obtained from the Chief, OMSAB, Mail Drop 80, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711 USA.

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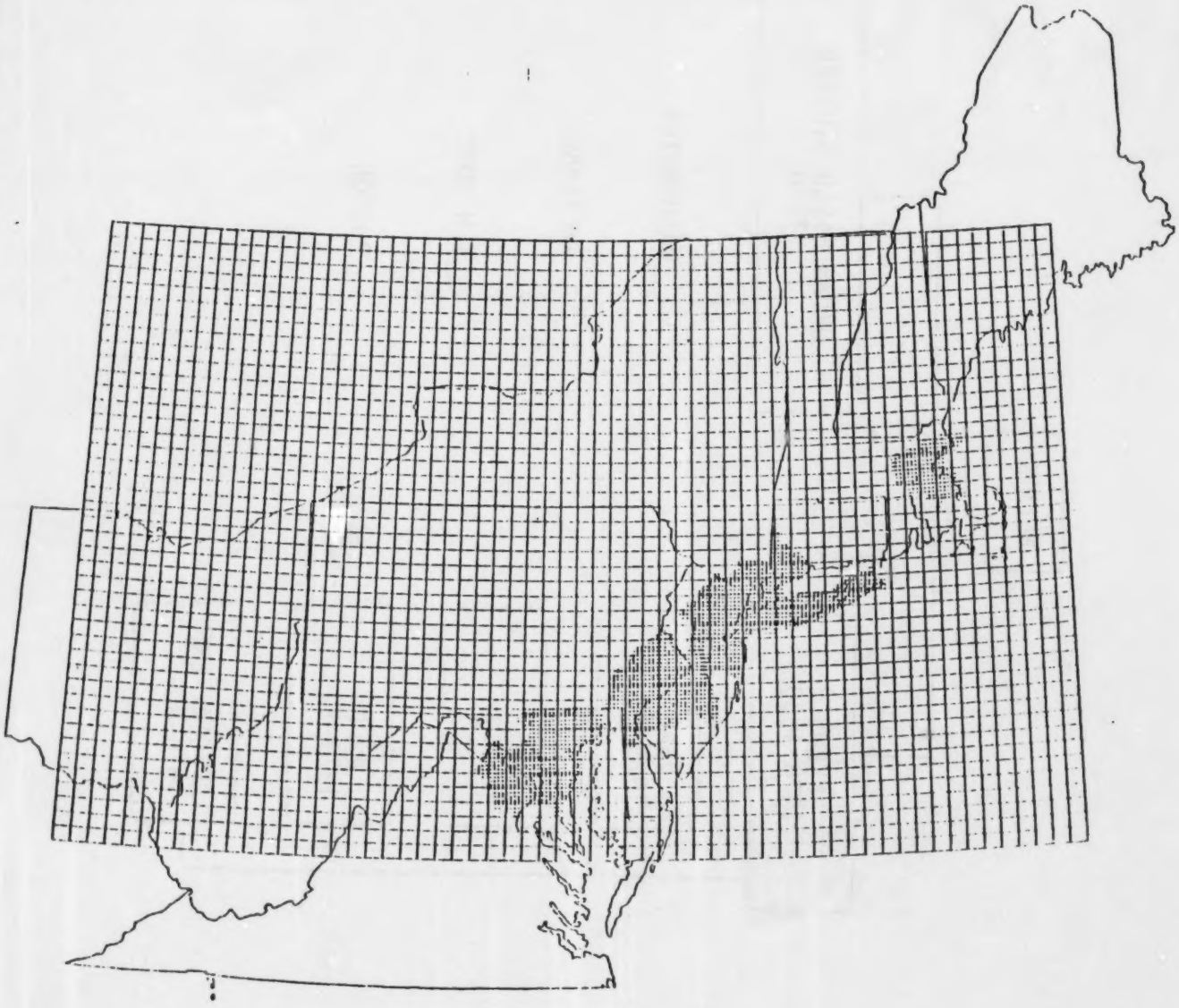

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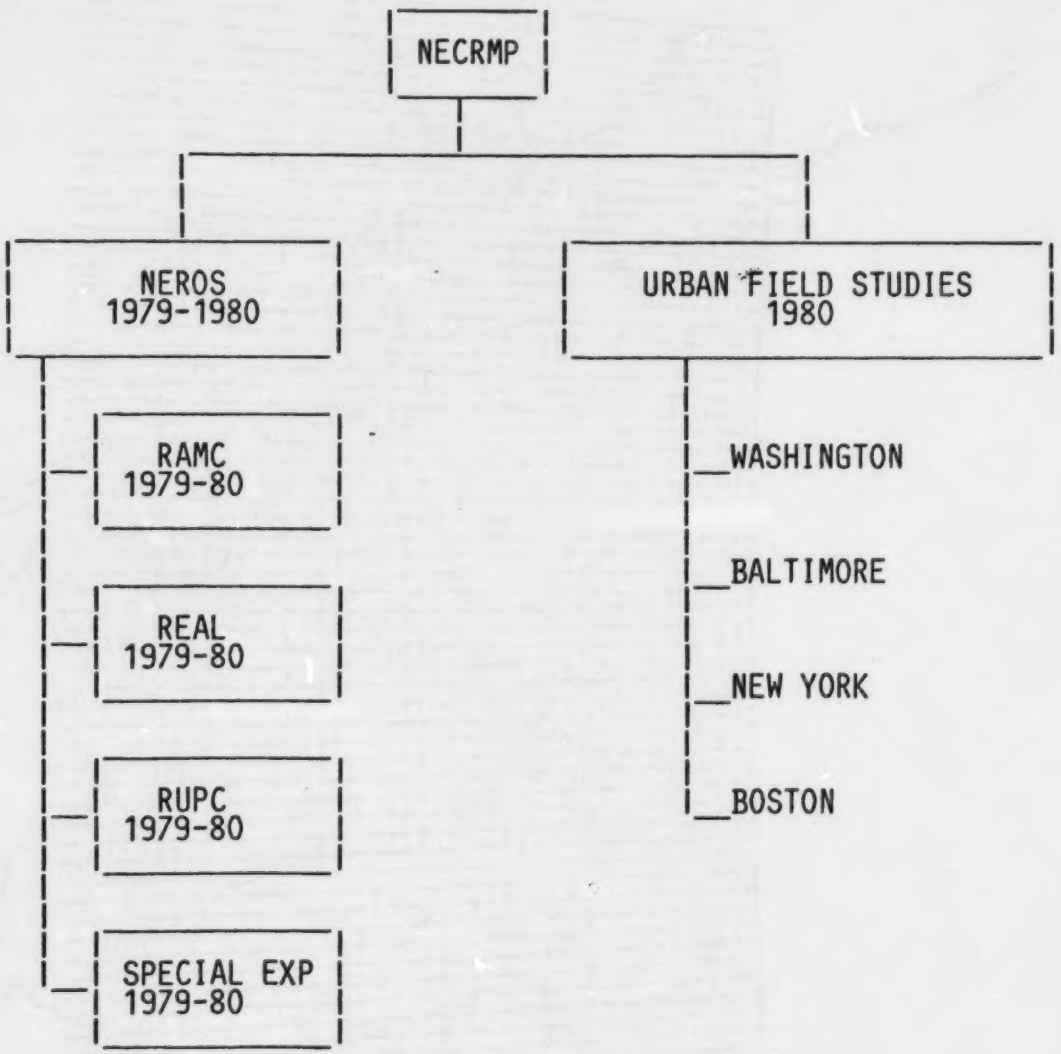

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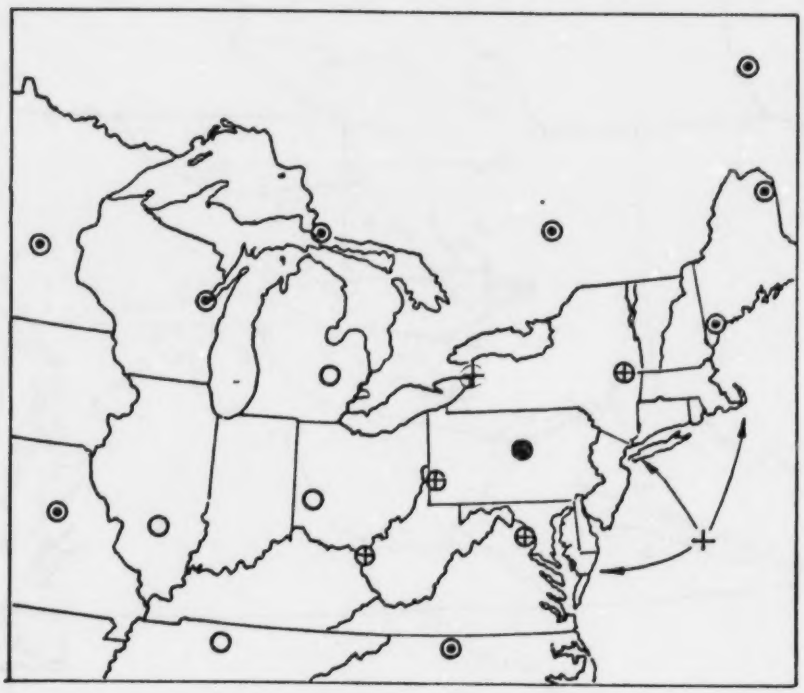

Fig. 3. Radiosonde stations near modeling area. The nine stations indicated by open circles took radiosondes at 6-h intervals during the 1980 NEROS; the eight stations indicated by a plus sign took radiosondes at 6-h intervals during the 1979 NEROS; the site in western Pennsylvania indicated by a solid circle was a special 6-h station established for the 1979-1980 NEROS programs.

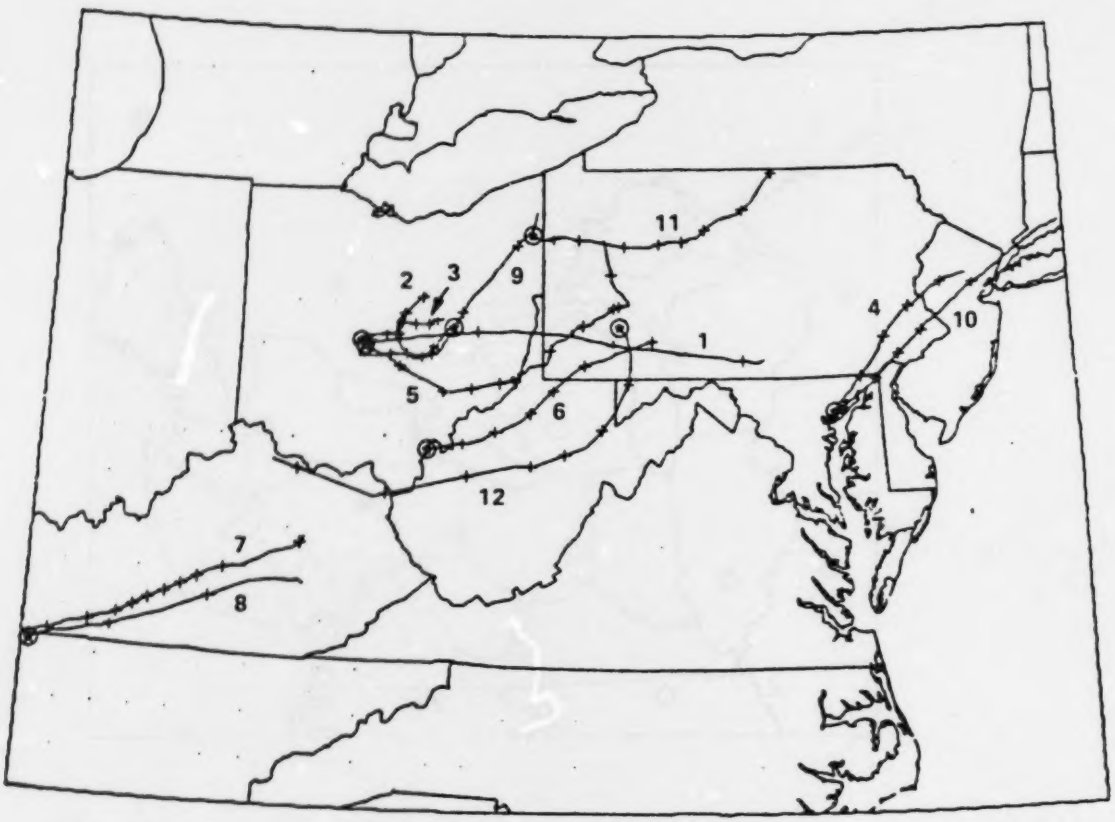

Fig. 4. Flight tracks of tetroons with positions every three h
marked by (+).

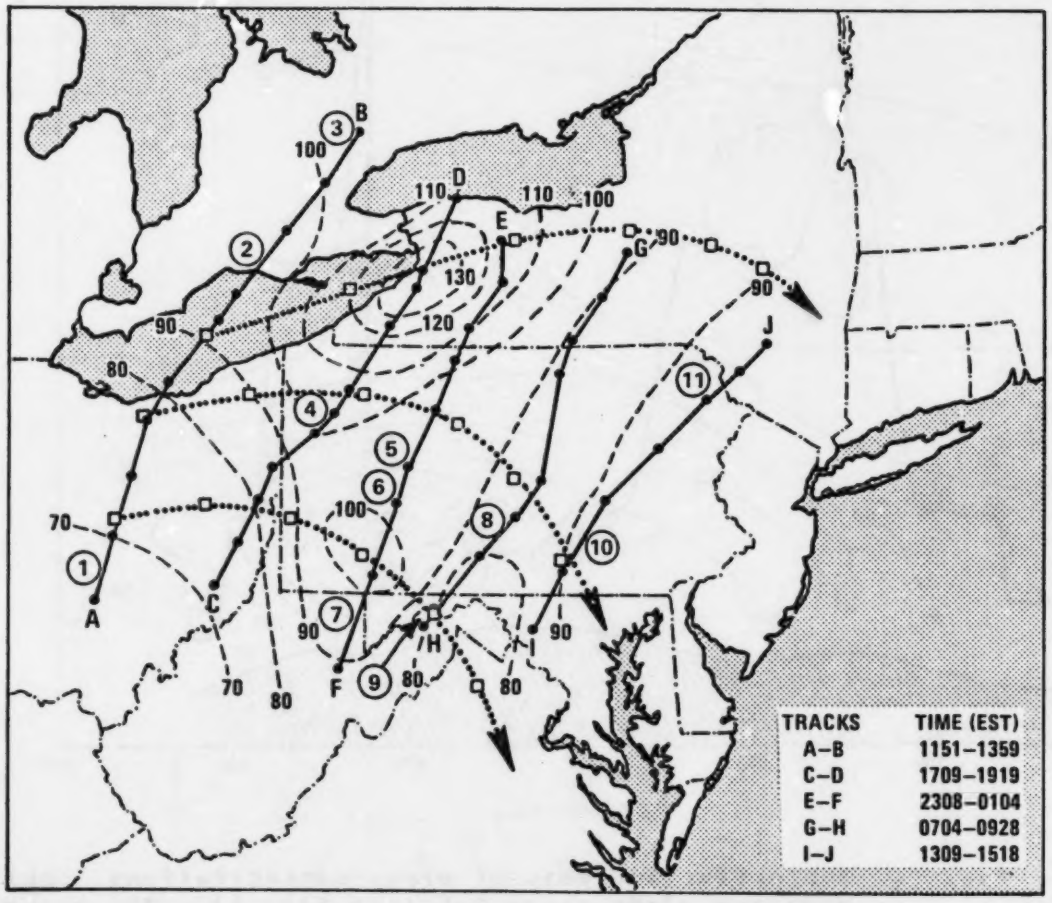

Fig. 5. Aircraft flight tracks, ozone concentrations (ppb) near the center of the mixed layer, and boundary layer trajectories initialized along track A-B at 1300 EST on 3 August 1979 (squares indicate 6-h positions). The circled numbers indicate the location of hydrocarbon samples.

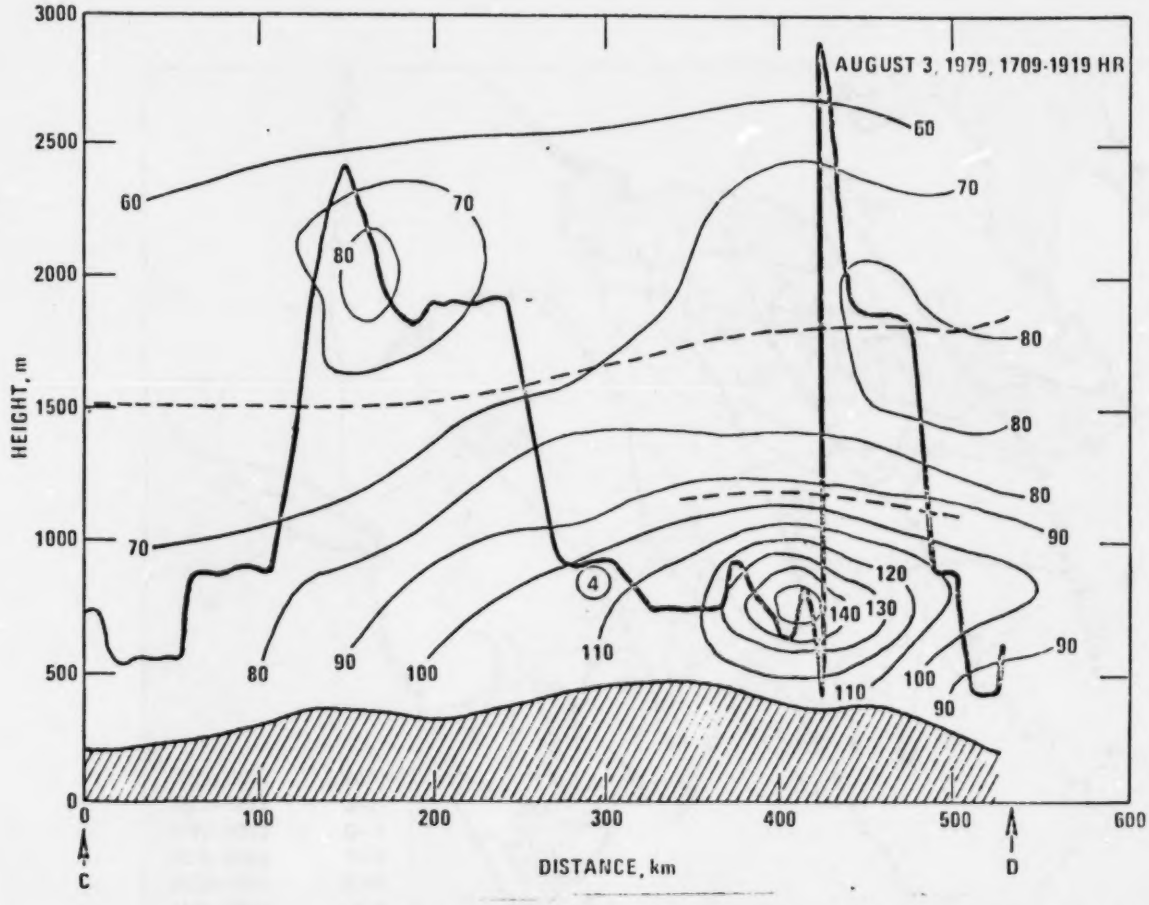

Fig. 6. Subjective analysis of ozone concentrations (ppb) measured along track C-D (see Fig. 5). The heavy solid lines indicate the aircraft flight path; the dashed lines indicate the top of the afternoon mixed layer; the circled number the location of a hydrocarbon sample.

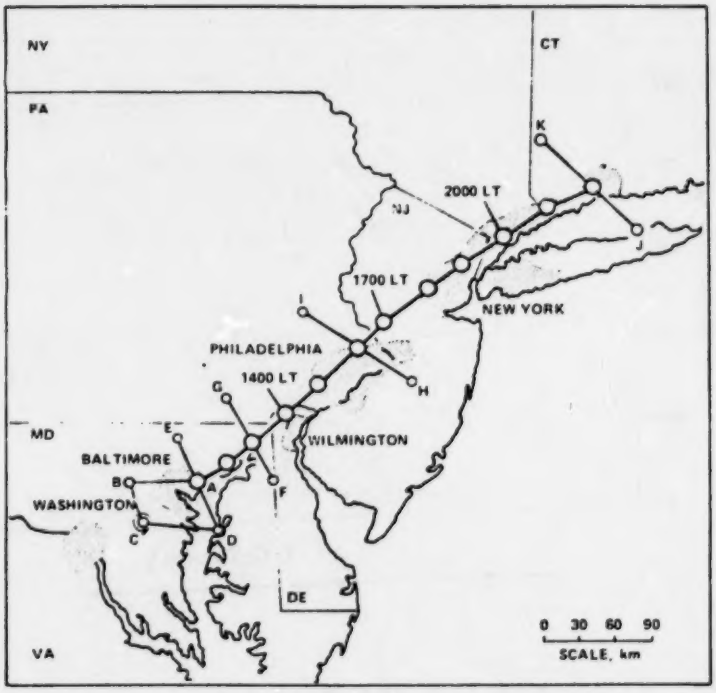

Fig. 7. Hourly location of tetroon (open circles) and airc
flight tracks on 14 August 1979.

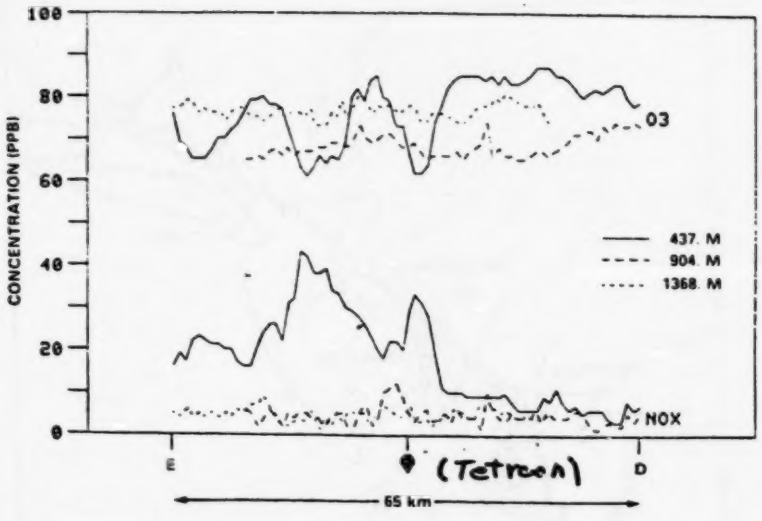

Fig. 8. Ozone and NOx concentrations along Lagrangian traverses D-E (see Fig. 7) between 0955 and 1041 EST on 14 August 1980. The location of the tetroon at 1000 EST is indicated at the bottom of the figure.

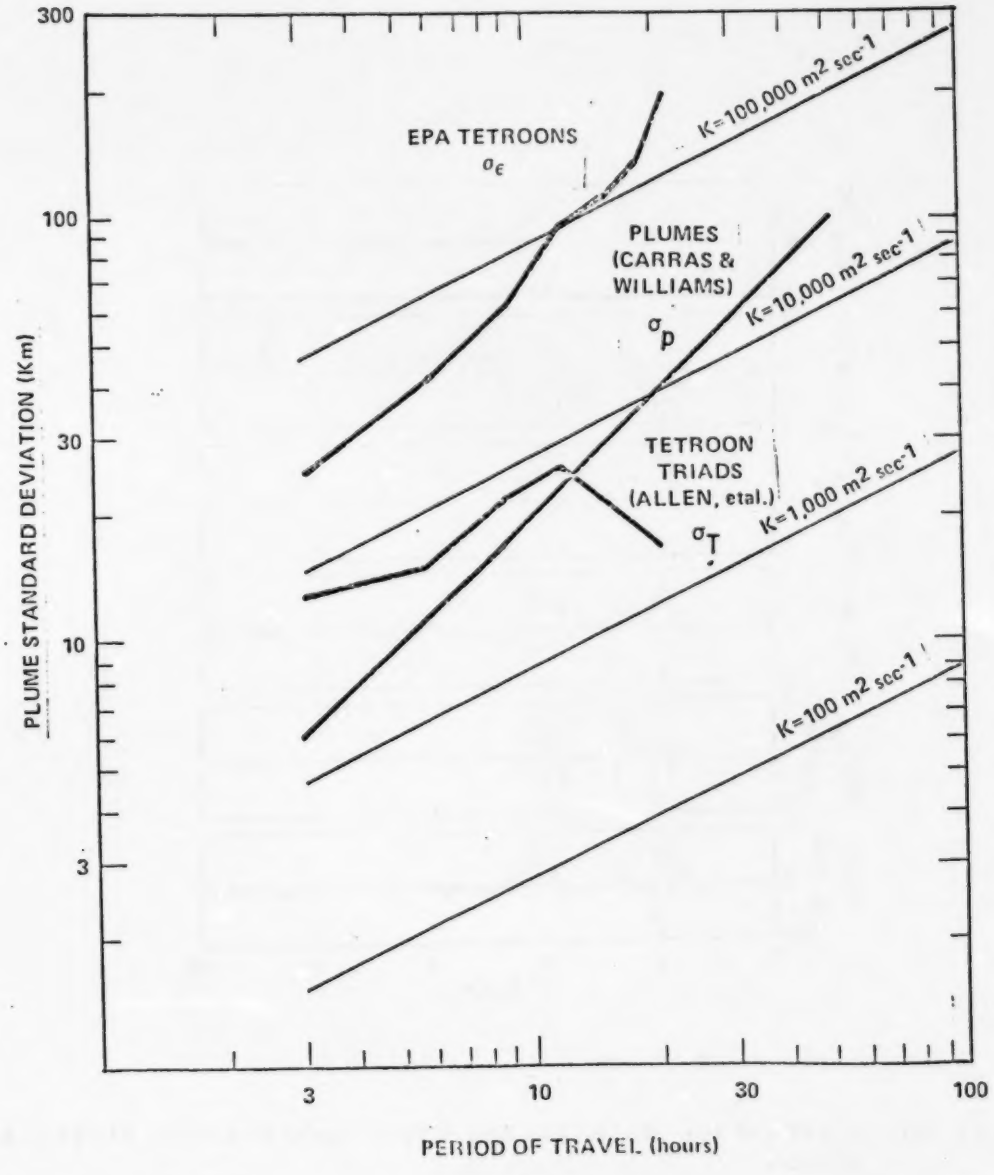

Fig. 9. Standard deviations of the differences between tetroon and diagnostic trajectories σ_ϵ, tetroon triad separation σ_T, and plume data σ_p.

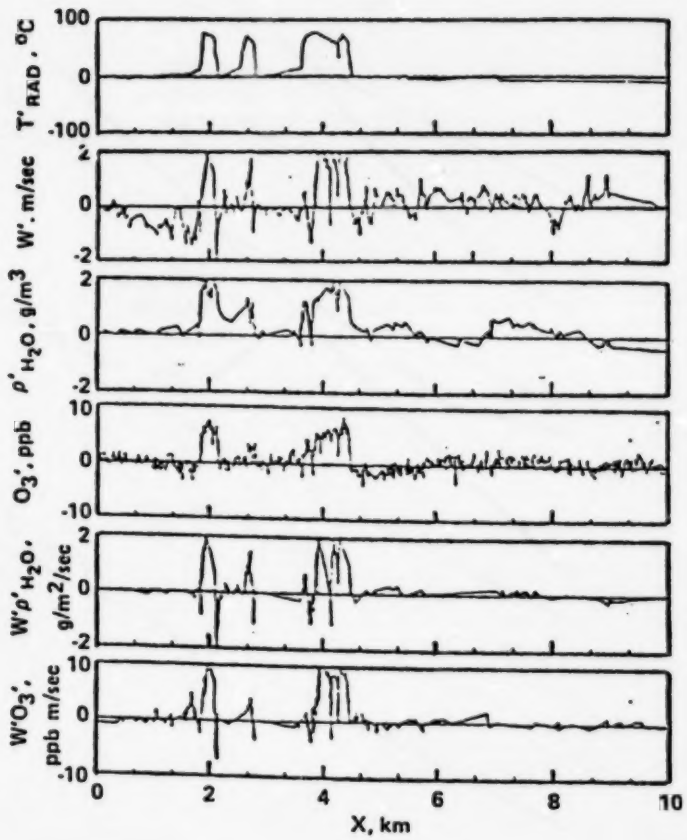

Fig. 10. Fluctuation data during cloud penetration flight in 1979 HEROS.

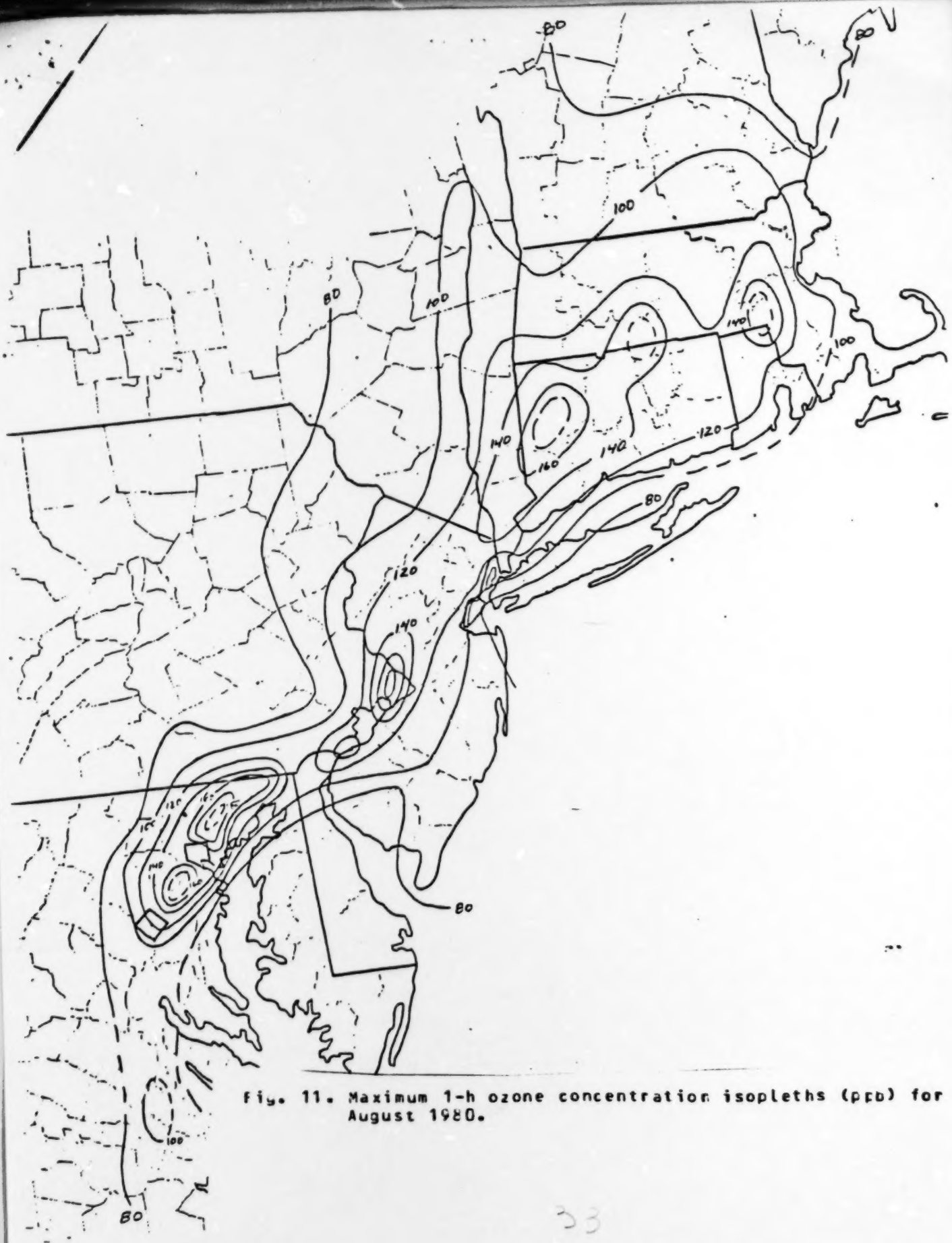

Fig. 11. Maximum 1-h ozone concentration isopleths (ppb) for 5 August 1980.